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Practitioner's Docket No.: 791_056 DIV

PATENT

ON APPEAL

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE BEFORE THE HONORABLE BOARD OF PATENT APPEALS AND INTERFERENCES

In re the application of: Hiroshi NEMOTO, Michio TAKAHASHI and Kenshin KITOH

Ser. No.: 10/071,664

Group Art Unit: 1745

Filed: February 8, 2002

Examiner: Raymond Alejandro

Confirmation No.: 4184

For: LITHIUM SECONDARY BATTERY

Mail Stop Appeal Brief-Patents
Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

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Janet M. Stevens

Janet M. Stevens

TRANSMITTAL OF BRIEF ON APPEAL

Sir:

Transmitted herewith are three copies of the Appellant's Brief on Appeal together with a check in the amount of $330.00 to cover the fee under 37 CFR 1.17(c).

The Commissioner is hereby authorized to charge any additional fees associated with this communication or credit any overpayment to Deposit Account No. 50-1446.

Respectfully submitted,
BURR & BROWN

Kevin C. Brown

June 21, 2004
Date

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Enclosures: Brief on Appeal (3)

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BRIEF ON APPEAL

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TABLE OF AUTHORITIES

I. REAL PARTY IN INTEREST

The real party in interest in the present appeal is NGK Insulators, Ltd., as reflected by an Assignment document recorded in the USPTO microfilm at reel 010101, frames 0816-0818.

II. RELATED APPEALS AND INTERFERENCES

There are no appeals or interferences regarding any application which is related to the present application.

III. STATUS OF CLAIMS

Claims 1 and 11-27 are pending.

Claims 14, 16 and 19-27 have been withdrawn from consideration by the USPTO.

Claims 2-10 have been canceled.

Claims 1, 11-13, 15, 17 and 18 stand finally rejected.

The applicants are appealing the rejection of claims 1, 11-13, 15, 17 and 18 (these claims are set forth in Appendix A attached hereto).

IV. STATUS OF AMENDMENTS FILED AFTER FINAL REJECTION

An Amendment after final rejection was filed on May 25, 2004 and has been entered in this application.

V. SUMMARY OF THE INVENTION AND THE APPLIED REFERENCES

The Invention

The present invention relates to a lithium secondary battery which has small internal resistance and has good charge-discharge cycle characteristics, with a lithium transition metal compound being used as a positive active material (specification, page 1, lines 4-9). Battery capacity as well as charge-discharge cycle characteristics ("cycle characteristics") in lithium secondary batteries heavily depend on the material characteristics of the positive active material to be used (specification, page 2, lines 5-8).

Conventionally, for improving electronic conductivity of a positive active material, attention was only paid to electronic conductivity among particles of a positive active material, but the relationship between diffusion of Li^+ and electronic conductivity inside a particle of a positive active material at the time of battery reaction was not regarded as a problem (specification, page 5, lines 2-7).

Detachment of Li^+ from a particle of a positive active material as well as insertion of Li^+ to a particle of a positive active material proceeds by diffusion of Li^+ inside a particle of a positive active material, simultaneously accompanied by transfer of electrons taking place inside a particle of a positive active material (specification, page 5, lines 8-12). At this time, if electronic conductivity inside a particle of a positive active material is low, diffusion of Li^+ hardly is apt to take place and velocity of detachment and insertion of Li^+, namely velocity of the battery reaction, becomes slow (specification, page 5, lines 12-16). The result is an increase in internal resistance, which has not been taken into consideration in the past (specification, page 5, lines 16-17). The present inventors paid attention to this point, and considered in earnest how to

improve electronic conductivity of a positive active material so that diffusion of Li^+ inside a positive active material may proceed well, thus reducing resistance of the positive active material (specification, page 5, lines 18-22).

In accordance with the present invention, there is provided a lithium secondary battery, comprising a lithium transition metal compound $LiMe_XO_Y$, in which a portion of transition element Me is substituted by not less than two additional elements to constitute $LiM_ZMe_{X-Z}O_Y$ (herein M represents substitution elements, and M ≠ Me, and Z represents quantity of substitution), the $LiM_ZMe_{X-Z}O_Y$ being used as a positive active material (specification, page 6, lines 4-11). As one of lithium transition metal compounds used in the present invention, lithium manganese oxide, especially a lithium manganese oxide having a spinel configuration of cubic system, may be employed (specification, page 7, lines 3-6). When $LiMn_2O_4$ is used, the substitution quantity Z preferably falls within a range of $0.01 \leq Z \leq 0.5$ (specification, page 18, lines 21-23). Examples of lithium transition metal compounds according to the present invention include $Li(Ni_{0.5}Ti_{0.5})_{0.01}Mn_{1.99}O_4$, $Li(Ni_{0.5}Ti_{0.5})_{0.1}Mn_{1.90}O_4$, $Li(Ni_{0.5}Ti_{0.5})_{0.15}Mn_{1.85}O_4$, $Li(Ni_{0.5}Ti_{0.5})_{0.3}Mn_{1.70}O_4$, and $Li(Ni_{0.5}Ti_{0.5})_{0.5}Mn_{1.50}O_4$ (specification, page 24, lines 11-15).

Starting raw materials were weighed and mixed, and then fired at 800°C in an air atmosphere for 24 hours, so that the positive active material compositions of $Li(Ni_{0.5}Ti_{0.5})_{0.01}Mn_{1.99}O_4$ and $Li(Ni_{0.5}Ti_{0.5})_{0.10}Mn_{1.90}O_4$ were obtained, and, for comparison, compositions of $Li(Co_{0.5}Ti_{0.5})_{0.02}Mn_{1.98}O_4$ and $Li(Co_{0.5}Ti_{0.5})_{0.10}Mn_{1.90}O_4$ were obtained (specification, page 23, lines 4-8 and 10-12; Declaration Under 37 CFR 1.132 filed December 24, 2003, page 2, lines 7-12).

Coin cells were prepared by mixing each of the respective positive active materials with

acetylene black powder as a conductive material and polyvinylidene fluoride as a bonding material in a ratio of 50 parts positive active material, 2 parts acetylene black powder and 3 parts polyvinylidene fluoride and press forming to prepare disc shapes which were incorporated in coin cells (specification, page 27, line 15 - page 28, line 3; Declaration Under 37 CFR 1.132 filed December 24, 2003, page 2, lines 13-23).

Internal resistance ratios of the respective coin cells were measured, and it was found that the internal resistance ratio of the coin cell having $Li(Ni_{0.5}Ti_{0.5})_{0.01}Mn_{1.99}O_4$ as its positive active material was 52%, the internal resistance ratio of the coin cell having $Li(Ni_{0.5}Ti_{0.5})_{0.10}Mn_{1.90}O_4$ as its positive active material was 36%, the internal resistance ratio of the coin cell having $Li(Co_{0.5}Ti_{0.5})_{0.02}Mn_{1.98}O_4$ as its positive active material was 96%, and the internal resistance ratio of the coin cell having $Li(Co_{0.5}Ti_{0.5})_{0.10}Mn_{1.90}O_4$ as its positive active material was 89% (specification, page 28, line 12 - page 29, line 6 and page 24, Table 1; Declaration Under 37 CFR 1.132 filed December 24, 2003, page 2, line 24 - page 3, line 8 and page 3, Table 1).

The Applied References

Manev '089

Manev '089 discloses a positive electrode material for lithium secondary cells which, according to the patent, exhibits good cycleability, reversible specific capacity, and structural stability (Manev '089, Abstract, lines 1-4). The positive electrode material comprises a lithium multi metal oxide having the general formula:

$$Li_{1+x}Mn_{2-Y}M_{m1}^{1}M_{m2}^{2} \ldots M_{mk}^{k}O_{4+Z}$$

wherein M^1, M^2, . . . M^k are at least two cations different than lithium or manganese, selected from the group consisting of alkaline earth metals, transition metals, B, Al, Si, Ga and Ge, X, Y, m_1, m_2, . . . m_k are numbers between 0 and 0.2; m_1, m_2 and Y are greater than 0; Z is a number between -0.1 and 0.2; and wherein the metals M^1, M^2, . . . M^k and the corresponding values m_1, m_2, . . . m_k satisfy the equation $Y = X + m_1 + m_2 + \ldots + m_k$ (Manev '089 Abstract, lines 4-18).

According to Manev '089, in a particularly preferred embodiment of the invention, the spinel compound is codoped with equivalent amounts of Co^{3+} and Ti^{4+} to form a spinel material having a composition described by the formula:

$$Li_{1+x}Mn_{2-X-2m}Co_m^{3+}Ti_m^{4+}O_{4+Z}$$

wherein X and m are molar parts with numbers between 0 and 0.2 and Z is a number between -0.1 and 0.2 (Manev '089, col. 2, lines 54-61).

Manev '089 discloses that although the codopant combination of cobalt and titanium is described as a preferred embodiment, various other combinations can be used (Manev '089, col. 4, lines 39-42). For example, Manev '089 discloses, combinations of aluminum, cobalt, chromium, copper, iron, gallium, magnesium, nickel, germanium, molybdenum, niobium, titanium, vanadium and tungsten can be used to produce multiple doped lithium manganese oxide spinels (Manev '089, col. 4, lines 42-45). Manev '089 lists a number of such combinations, including aluminum/titanium, gallium/titanium, nickel/titanium, iron/titanium, chromium/titanium, cobalt/vanadium, aluminum/vanadium, magnesium/vanadium, gallium/vanadium, nickel/vanadium, iron/vanadium, chromium/vanadium, cobalt/molybdenum,

aluminum/molybdenum, gallium/molybdenum, nickel/molybdenum, iron/molybdenum, chromium/molybdenum, cobalt/germanium, aluminum/germanium, magnesium/germanium, gallium/germanium, nickel/germanium, iron/germanium, chromium/germanium, cobalt/nickel/vanadium, magnesium/germanium/vanadium, aluminum/cobalt/titanium, aluminum/titanium/molybdenum, aluminum/cobalt/molybdenum, nickel/titanium/molybdenum, cobalt/nickel/titanium/vanadium, cobalt/nickel/titanium/molybdenum, and cobalt/nickel/aluminum/titanium/vanadium (Manev '089, col. 4, lines 45-60).

Manev '089 discloses two examples, in both of which a codopant combination of equal amounts of cobalt and titanium was employed (Manev '089, col. 7, lines 18-60).

According to Manev '089, the multiple-doped lithium manganese oxide spinel compounds can be used in positive electrodes in secondary lithium and lithium-ion cells by placing such a positive electrode inside a secondary lithium or lithium-ion cell with a lithium counterelectrode and an electrolyte (Manev '089, col. 7, lines 1-15).

Biensan '645

Biensan '645 is directed to an electrode for a rechargeable lithium cell, containing an electro-chemically active material with general formula $Li_xM_yA_mD_zO_t$, where: $0.8 \leq x \leq 1.2$; $0 \leq z \leq 0.3$; $1.8 \leq t \leq 4.2$; $(0.8 - m - z) \leq y \leq (2.2 - m - z)$; $0 < m \leq 0.3$; where M is at least one transition metal selected from nickel, cobalt, manganese, and iron, A is selected from magnesium and calcium, and D is at least one element selected from the elements of groups 4b to 5a of the periodic table of elements (Biensan '645, Abstract). Biensan '645 also discloses a rechargeable lithium cell comprising such an electrode and a negative electrode which comprises

an electro-chemically active material selected from lithium metal, lithium alloys and a material which can reversibly intercalate lithium ions into its structure, such as carbons (graphite, vitreous carbon, coke) in the form of a powder or fibers, or metal oxides (Biensan '645, col. 2, line 65 - col. 3, line 6).

VI. ISSUE

1. Whether appealed claims 1, 11-13, 15, 17 and 18 would have been obvious to one of ordinary skill in the art in view of U.S. Patent No. 6,040,089 (Manev '089) in view of U.S. Patent No. 6,071,645 (Biensan '645).

VII. GROUPING OF CLAIMS

Each pending claim of the present application is separately patentable, and upon issuance, shall be presumed valid independently of the validity of the other claims. 35 USC §282.

The patentability of claims 1, 11-13, 15, 17 and 18 is addressed together herein, and so for purposes of 37 C.F.R. 1.192 (c)(7), claims 1, 11-13, 15, 17 and 18 will stand or fall together.

VIII. ARGUMENT

In view of the following, appealed claims 1, 11-13, 15, 17 and 18 would not have been obvious to one of ordinary skill in the art in view of U.S. Patent No. 6,040,089 (Manev '089) in view of U.S. Patent No. 6,071,645 (Biensan '645).

The present invention is directed to a lithium secondary battery comprising a negative active material including carbon, and a positive active material including a lithium transition

metal compound. In accordance with the present invention, the lithium transition metal compound is represented by the formula $Li(Ni_{X1}Ti_{X2})_Z Mn_{2-Z}O_4$ wherein z is 0.01 to 0.5, $X_1 > 0$, $X_2 > 0$, $X_1+X_2=1$.

As noted above, Manev '089 discloses a positive electrode material which comprises a lithium multi metal oxide having the general formula:

$$Li_{1+x}Mn_{2-Y}M_{m1}{}^{1}M_{m2}{}^{2}\ldots M_{mk}{}^{k}O_{4+Z}$$

wherein M^1, M^2, . . . M^k are at least two cations different than lithium or manganese, selected from the group consisting of alkaline earth metals, transition metals, B, Al, Si, Ga and Ge. Throughout its disclosure, Manev '089 emphasizes and focuses on cobalt and titanium as co-dopants. According to Manev '089, in a particularly preferred embodiment of the invention, the spinel compound is co-doped with equivalent amounts of Co^{3+} and Ti^{4+} to form a spinel material having a composition described by the formula:

$$Li_{1+x}Mn_{2-X-2m}Co_m{}^{3+}Ti_m{}^{4+}O_{4+Z}$$

Manev '089 discloses two examples, in both of which a co-dopant combination of equal amounts of cobalt and titanium was employed (Manev '089, col. 7, lines 18-60).

Other than the extremely broad disclosure in col. 4, lines 39-62, which encompasses 16,369 possible combinations, including 91 possible two-component combinations and 364 possible three-component combinations, and lists thirty-four combinations (among them being

"nickel/titanium") nowhere does Manev '089 provide disclosure regarding any other co-dopants, or examples which employed other co-dopants (let alone use of nickel and titanium as co-dopants). Manev '089 contains no disclosure regarding any expectation of favorable properties of the listed combinations, relative to any of the other 16,369 encompassed combinations.

Based on the disclosure in Manev '089, one of ordinary skill in the art would expect oxide cathode material which is multiple-doped with each of the combinations disclosed in Manev '089 to have properties which are at best (in view of the clear preference in Manev '089 for a titanium/cobalt co-dopant) similar to the properties obtained with oxide cathode material which is multiple-doped with titanium and cobalt, the combination described throughout Manev '089 as the preferred combination and the only combination regarding which Manev '089 contains disclosure of actually making or testing.

The present applicants have demonstrated that *completely contrary to such expectation*, oxide cathode material which is multiple-doped with nickel and titanium ***do not*** have properties which are merely similar at best to the properties obtained with oxide cathode material which is multiple-doped with titanium and cobalt.

That which would have been surprising to a person of ordinary skill in a particular art would not have been obvious. *In re Soni*, 34 USPQ 2d 1684, 1687 (Fed. Cir. 1995). This principle applies most often to the less predictable fields, such as chemistry, where minor changes in a product or process may yield substantially different results. *In re Soni*, 34 USPQ 2d at 1687.

As noted above, positive active material compositions of $Li(Ni_{0.5}Ti_{0.5})_{0.01}Mn_{1.99}O_4$ and $Li(Ni_{0.5}Ti_{0.5})_{0.10}Mn_{1.90}O_4$ were obtained, and, for comparison, compositions of

$Li(Co_{0.5}Ti_{0.5})_{0.02}Mn_{1.98}O_4$ and $Li(Co_{0.5}Ti_{0.5})_{0.10}Mn_{1.90}O_4$ were obtained. The internal resistance ratio of the coin cell having $Li(Ni_{0.5}Ti_{0.5})_{0.01}Mn_{1.99}O_4$ as its positive active material was found to be *52%* and the internal resistance ratio of the coin cell having $Li(Ni_{0.5}Ti_{0.5})_{0.10}Mn_{1.90}O_4$ as its positive active material was found to be *36%*, while the internal resistance ratio of the coin cell having $Li(Co_{0.5}Ti_{0.5})_{0.02}Mn_{1.98}O_4$ as its positive active material was found to be *96%*, and the internal resistance ratio of the coin cell having $Li(Co_{0.5}Ti_{0.5})_{0.10}Mn_{1.90}O_4$ as its positive active material was found to be *89%* (i.e., *more than double* the value where an equal amount of nickel/titanium co-dopant was used instead of cobalt/titanium co-dopant).

It is respectfully submitted that the above-described comparative testing dispels any notion that a nickel/titanium co-dopant would behave in a manner which is similar to the manner in which a titanium/cobalt co-dopant behaves, thereby rendering the present claims non-obvious in view of the applied combination of references. Moreover, the comparative testing demonstrates that the claimed subject matter achieves results which are unexpectedly superior to those obtained in accordance with the use of a titanium and cobalt co-dopant.

In view of the above, reversal of the rejection of claims 1, 11-13, 15, 17 and 18 under 35 U.S.C. 103(a) over Manev '089 in view of Biensan '645 is respectfully requested.

IX. CONCLUSION

Accordingly, the Honorable Board of Patent Appeals and Interferences is respectfully requested to reverse the Final Rejection of claims 1, 11-13, 15, 17 and 18 under 35 U.S.C. 103(a) over Manev '089 in view of Biensan '645.

The Commissioner is hereby authorized to charge any additional fees associated with this communication or credit any overpayment to Deposit Account No. 50-1446.

Respectfully submitted,

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June 21, 2004
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APPENDIX A

1. A lithium secondary battery comprising a negative active material including carbon, and a positive active material including a lithium transition metal compound, said compound being represented by the formula $Li(Ni_{X1}Ti_{X2})_ZMn_{2-Z}O_4$ wherein z is 0.01 to 0.5, $X_1 > 0$, $X_2 > 0$, X1+X2=1, and said positive active material has a spinel configuration of the cubic system.

11. A lithium secondary battery according to Claim 1, wherein said lithium transition metal compound is selected from the group consisting of $Li(Ni_{0.5}Ti_{0.5})_{0.01}Mn_{1.99}O_4$, $Li(Ni_{0.5}Ti_{0.5})_{0.1}Mn_{1.90}O_4$, $Li(Ni_{0.5}Ti_{0.5})_{0.15}Mn_{1.85}O_4$, $Li(Ni_{0.5}Ti_{0.5})_{0.3}Mn_{1.70}O_4$, and $Li(Ni_{0.5}Ti_{0.5})_{0.5}Mn_{1.50}O_4$.

12. A lithium secondary battery according to Claim 1, wherein said lithium transition metal compound further comprises Mg as an additional element.

13. A lithium secondary battery according to Claim 1, wherein said lithium transition metal compound further comprises Li as an additional element.

15. The lithium secondary battery of Claim 1, wherein the average ionic radius of the substitution members is within ± 15 percent of the ionic radius of Mn.

17. The lithium secondary battery according to Claim 1, wherein the lithium transition metal compound is composed by firing a mixed compound comprising salts and/or oxides having been prepared with a predetermined ratio in the presence of oxygen within a temperature range of 600°C to 1000°C for 5 hours to 50 hours.

18. The lithium secondary battery according to Claim 17, wherein the lithium transition metal compound has been synthesized and obtained by conducting at least first and second firing steps, with the firing temperature of the second step being higher than that of the first step.